Report on Westlake Landfill Phytoforensic Assessment using Gamma Spectroscopy

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Summary

The Missouri University of Science and Technology was contacted by Missouri Attorney General's Office with a request to assess potential contamination in the area of West Lake Landfill (WLL). The landfill has two areas; Radiological Area 1 and 2 which contain radioactive material. Dr. Shoaib Usman contributed to this site assessment endeavor by performing gamma spectroscopy of the tree samples for any trace amounts of radioactivity. As part of this effort, historical data from the site was evaluated. It was found that the previous reports have indicated the presence of certain radioactive materials in the soil and water samples. In this study, additional plant sampling was conducted for these pollutants using phytoforensics. While the uptake of heavy metal and radioactive materials in plants have been reported by others, no such study is available for the WLL site to the best of the author's knowledge.

Gamma spectroscopy of the samples indicates that there are trace amounts of ^{238}U, ^{235}U and ^{232}Th and their respective daughter products in some samples of the tree cores. These traces are seen in clusters, meaning there are areas with a number of trees containing the material and other areas showing no sign of these materials. Based on the statistical analysis, it is likely that the soil and/or water at the root tip of these trees also contain the radioactive material. This study is for screening purposes only, therefore no quantitative data on radioactive material per unit tree core mass is available at this time.

History of Westlake Landfill Site

The West Lake Landfill (WLL) has been in operation since the early 1950s. Due to the evolutionary nature of the environmental protection laws, the earlier procedures and methods are not well documented as it was not required at that time. Moreover, some of those methods may not be in agreement with today's standards and regulations. WLL history is documented elsewhere (1,2,3,4) and is not repeated here. Nevertheless, there is documentary evidence that certain radioactive materials were deposited in the landfill and can potentially be leaching to reach off-site locations. The purpose of this report is to summarize the findings of a screening study using phytoforensics method.

Vegetation Sampling and Phytoforensics Applications

While tree and plants are known to preferentially accumulate metals and pollutants from the environment (5, 6,7), to the best of the author's knowledge, no such study has been previously conducted at WLL site to investigate the possible leaching of waste material from the site. Recently, Missouri S&T received a request from Missouri Attorney General's Office to perform sampling of vegetation and phytoforensic testing to investigate any traces of contaminants in the plants. The effort was divided into two parallel analyses; organic chemical and radiological. The author was responsible for the radiological investigation of the plant samples. While there is evidence that certain plants do preferentially accumulate metals and radioactive material, at this time the author has no precise knowledge of the effectiveness of the plants sampled from the site in accumulating the radioactive traces found in them.

Sampling and Analytic Methods

Selection of trees for sampling was rather random, but due consideration given to; the tree species, and trunk diameter and spatial coverage of the WLL area. The location of each tree sampled was recorded using a Global Positioning System (GPS). Pictures of selected sampled trees were taken to identify the tree species. Accessibility of the tree and wide geographic coverage surrounding the waste site was also considered in tree selection.

Missouri S&T has significant experience in plant sampling methods; using an increment borer (8), increment hammer (9), and branch sampling method (10). Simonich and Hites have reported bark sampling method's success (11,12). All in all these techniques are well accepted by the scientific community.

The core samples were taken on April 23-24, 2015 by Missouri S&T students under the guidance of Dr. Joel G. Burken, from the West Lake Landfill (WLL) site, while Dr. Shoaib Usman was present on April 24th for the sampling. Subsequently, a second sampling was performed on August 12, 2015 by Dr. Burken to confirm samples collected along Old St. Charles Road.

Tree cores were collected at chest height (approximately 1.5 m from the ground) using a 0.5-cm increment borer (Forestry Services Inc., Pawleys Island, SC). The cores were approximately 8 cm in length and immediately after extraction, the samples were transferred to a 20-mL vial with a screw-top cap and Teflon/silicone septa (Supelco, Bellefonte, PA). For the chemical analysis these vials are important to retain all the organics; for the radiological analysis the vials offered a means to prevent any cross contamination between the samples and the surroundings. Field blanks with air samples were taken every 10 to 15 samples. After the sampling was completed, the vials were stored on ice and then transferred directly to Missouri S&T laboratories for refrigerated storage and subsequent analysis.

Batching for Wider Screening

A large number of tree samples were available for analysis. In order to screen samples in a timely manner and yet offer sufficient insight into the geographic variability, 10 samples were grouped together to form a "Batch." These batches were counted (10 tree samples simultaneously) to efficiently covered a larger geographic area for contamination screening. Samples were arranged equally spaced from the detector as shown in Figure 3 of the Analytical report (2). The initial batch of 10 cores was counted for 24 hours. To improve the statistics the second batch was counted for 70 hours. All subsequent batches were counted for 50 hours as a compromise between time efficiency and counting statistics. All samples counts were time normalized to 100 hours for comparison with the background data, analysis and presentation.

Screening Methodology

Cores were also analyzed for radioactive contamination utilizing a High Purity Germanium (HPGe) Detector. The gamma counting and spectroscopy system was calibrated using standard sources of Cobalt (Co-60), Manganese (Mn-54), and Cesium (Cs-137). No efficiency calibration was possible due to the complex detector sample geometry and consequently no quantitative radioactivity contents in the samples were obtained. The counting system includes standard Multi-Channel

Analyzer (MCA) coupled with ORTEC MAESTRO Software for gamma spectroscopy. Energy Calibration curve is shown in Figure 2 of the Analytical report (2).

All measurements were taken in 217 Fulton Hall, which is a low background room lined with lead sheets on the walls and the floor. To assess the radiation background, a 100 hours background data without any source or sample was collected. This background spectrum was subtracted from each source or sample measurement to obtain the net counts or contribution due to the tree sample.

Statistical Nature of Radioactivity and 3σ Approach Rationale

When a sample is drawn from a population and a mean is calculated to represent the **population mean**, there is always a chance that a second sample from the same population will produce a mean which is different from the first mean. This statistical uncertainty is well established (13,14). It is well established that if only one sample is collected from a normally distributed quantity the probability that the next sample will be within one standard deviation (σ) of the first measurement is 68.27%, while the probability for it to be within two σ range is 95.45% and three σ range 99.73%.

As a rule of thumb, all batches with gross counts (sum of the entire spectrum) lower than the background plus three standard deviation (3σ), were considered as clean/free of contamination and were not analyzed any further. This choice of background plus 3σ is based on low probability of only 0.27% that the sample was a false positive and the higher counts were in fact due to the statistics uncertainty in the background. In other words, if a sample is found to have higher radiation counts than the background by more the 3σ, one can conclude that it is 99.73% probable that the sample contain some radioactive material beyond the background. Sample with total counts higher than the total background plus 3σ counts were analyzed further for possible contamination from Uranium and Thorium.

Based on the site history and the results from previous studies (4,15) uranium is a likely contaminant at the site. Again for each significant peak we followed the 3σ rule. If the count under the specific energy were at least background+3σ, we considered the specific peak/gamma present in the tree sample, otherwise the tree sample was considered clean from the specific isotope.

Results of Gamma Spectroscopy

Detailed results of our gamma spectroscopy are included in the Analytical report (2). As presented in Figures 5-7 of the report, most of the batches (11/16) of ten samples resulted in counts below the 3σ definition as clean, i.e., without any significant counts above background+3σ rule. However, five batches (batch number; 1, 3, 4, 5, 6) had exhibited counts where the gamma counts were statistically higher than the background, warranting further investigation. It turns out that these tree samples with higher than background (background plus 3σ or more) counts formed four clusters; North-West cluster i.e., at the top of Radiological Area 2, North-East cluster near Radiological Area 1, South-West cluster and the South-East cluster. The samples right from the top of the Radiological Area 1 showed no traces of U238, U25 and/or Th232. U238 counts were higher than the U235 while Th232 counts were smaller than both the U238 and U235. Two of these clusters; South-West cluster and the South-East were unexpected due to the distance between

those clusters and the known locations of radiological waste disposal. This rather surprising observation could have been due to the batching because if a batch is found to have higher than background counts, it means one or more (not necessarily all) of the tree samples contain radioactive material higher than the statistical nature of the counting. Nevertheless, this finding required a follow up measurement. A new batch of 12 tree samples from these two cluster location was counted again and we observed higher than background counts for this new batch as can be seen in figures 13a-c of the Analytical report (2).

Summary of Findings and Expert Opinion

Based on the data available after this study, one can conclude that there is 99.73% probability that one or more of the sample in batches 1, 3, 4, 5, 6 contain radioactive material. Four clusters were observed (see figure 1); the North-West cluster is at the top of Radiological Area 2 suggesting that the material may have migrated to reach the root tip of the plants right above. However, the samples the Radiological Area 1 showed no traces of U238, U235 and/or Th232. This observation suggests that the top soil and the tree root tips may be in geological isolation of the underlying radioactive material in the area. In other words, the tools and techniques put in place to prevent upward migration of the radioactive material seem to perform well to prevent upward migration and limit availability.

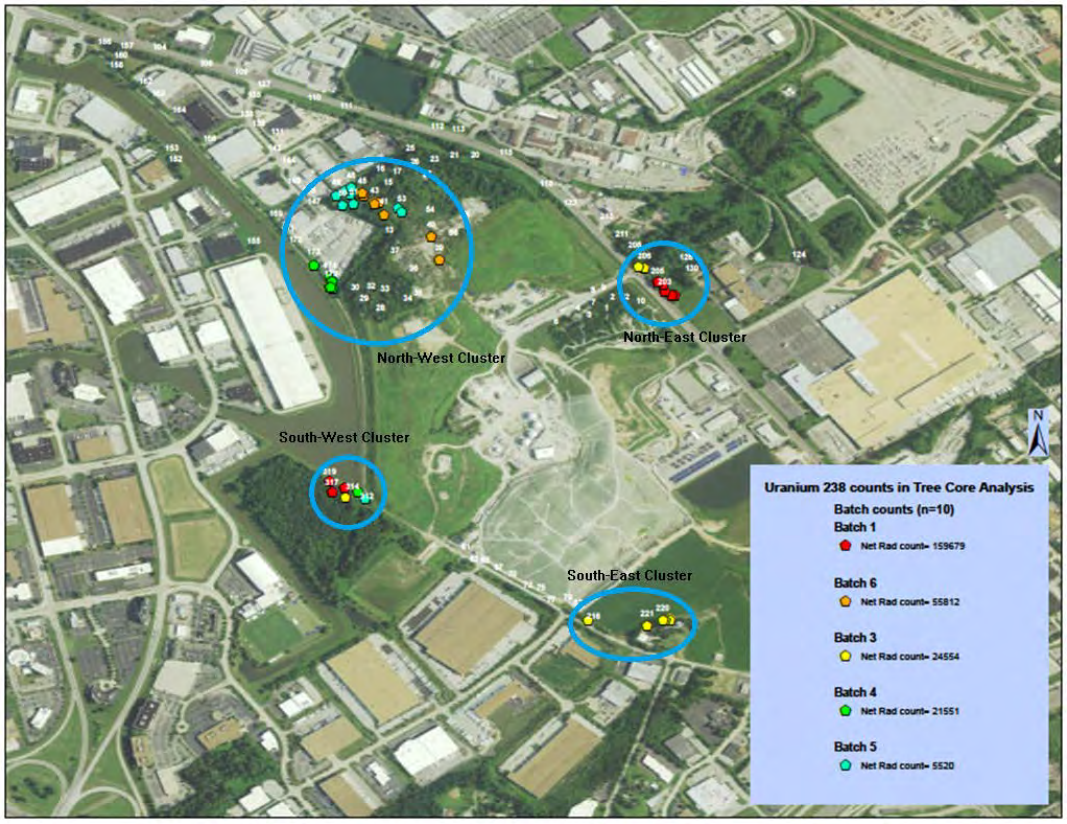

Figure 1: Clusters of Radioactive Material Observed at WLL

The second cluster; North-East cluster also showed counts higher than background (background plus 3σ or more) total counts and specific peaks significantly higher than the corresponding peak (more than 3σ) in the background. This finding suggested that the North-East cluster area which is in close proximity of the Radiological Area 1 could have received some radioactive material due to migration. The source of this radioactive material could potentially be the leached material migrated underground to this sampling area.

The third and fourth clusters were rather unexpected, therefore these measurements were repeated and the new batch with only tree samples from this cluster area (South-West and South-East) came positive outside the statistical uncertainty (3σ rule). Based on this observation one can conclude that one or more of the tree samples in this batch(es) was/were radioactive. Because of the batching we are unable to pin point the source of this possible contamination.

As stated earlier, no quantitative analysis is available nor the characteristics of the plantation in area is known to the author but qualitatively speaking, there is sufficient merit in the data to warrant careful analysis and in depth examination.

More work would be needed to analyze each sample individually to develop a more detailed understanding of the geographic distribution of the contamination but at this point the screen data shows pockets or clusters of elevated radioactivity in the tree samples. Two of the four clusters are in close proximity of the known Radiological areas but two other sites appear to contain radioactive material in the tree core. These locations are not in close proximity of the radiological areas suggesting possible movement of the material. Detailed investigation is recommended for thorough understanding of the source and transport mechanism of the radioactive material at the site.

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